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An Evaluation on Mixture Materials Using Overburden and Flyash as Cover Layer for Acid Mine Drainage Prevention and Underlying Materials of Seedbed in Indonesian Coal Mine

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Abstract: Almost of the coal produced is from open-cut mines in Indonesia. Mining operation of open-cut mines causes a decreasing of a tropical rainforest and gives serious impacts on surrounding environment. Therefore, the environmental reconstruction by revegetation in the disturbed land is required. Mining operation of an open-cut mines requires removal of overburden and placement of them as waste rock dump. However, some of overburden contains sulphide contents which causes Acid Mine Drainage (AMD) and may be undesirable for plant growth because of high salinity, high soluble metal concentrations and high acidity. Furthermore, additional of flyash to waste rock dump for neutralizing AMD is discussed in current researches. As the important object of rehabilitation program is the creation of stable and self-sustaining land surface by vegetation, an assessment of overburden and flyash as planting base have to be discussed. In this study, several geochemical static and kinetic tests for overburden and flyash were carried out to prevent AMD generation and to discuss the impact to plant growth. It was revealed that the adverse effect to plant growth from overburden and flyash can be reduced by effective AMD prevention and preparing appropriate curing period of the ground until the of dissolution of metal ions are decreased.

Keywords: Acid mine drainage, coal mining, cover layer system, flyash, rehabilitation

INTRODUCTION

Indonesia produces over 250 Mt of clean coal in 2011 and is the second largest coal exporter to Japan, accounting for 30 Mt/y. Over 99% of clean coal is produced from open-cut mines in Indonesia. As the demand of coal not only in Indonesia but also in the rest of the world is increased dramatically because of increasing the energy supply in the world, the development of coal mining is advanced in order to meet the demand of coal. However, it can be predicted that some serious impacts on surrounding environment occur with a mining operation of open-cut mines. Therefore, an appropriate rehabilitation program has to be considered and introduced for an environment protection.

The large scale development of open-cut mines in Indonesia causes a decreasing of a tropical rainforest. Now, the government and local residents show deep interest in an environmental protection. Accordingly, mining companies have to pay more attentions on the rehabilitation program after the open-cut mining operation has finished. Therefore, it is very important to consider the rehabilitation program in the initial stage when the mine plan is designed (Shimada *et al*., 2006). Rehabilitation has to be considered from perspectives, an environmental protection and an environmental reconstruction. If rehabilitation is conducted adequately in post mine surface area, it is possible to restore almost the original environment and ecosystem before development of open-cut mining.

Fig. 1: Acid mine drainage in Indonesia

Acid Mine Drainage (AMD) is considered as one of the most significant environmental issues in most coal surface mines. Indonesia belongs to rainforest climate and there are waste rocks contained sulphide contents. Therefore, AMD is easy to generate by oxidation of sulfide minerals contained in rocks by reaction with oxygen and water in Indonesia (Fig. 1). Some significant impacts on surrounding environment such as high acidity, high soluble metal concentrations and high sulfate/salinity can be estimated due to AMD generation. In order to achieve the success of rehabilitation in the post mined area, the appropriate management of acidic water has to be needed. As one of the efficient methods for AMD prevention, utilization of flyash (FA) is discussed in current researches. In general, FA has the alkaline properties even though its chemical and physical characteristics

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depend on the coal quality and combustion conditions. Having considered the high capacity potential for acid neutralizing due to its alkaline property, FA can be expected to be an effective solution for AMD prevention when the enough amount of NAF is difficult to prepare *in-situ*. However, the leaching of heavy metals from FA has to be taken into consideration. Moreover, the important object of rehabilitation program is the environmental reconstruction and the creation of stable and self-sustaining land surface by vegetation. Therefore, the effect for plant growth from AMD and FA has to be considered.

This paper describes the AMD generation mechanism and its prevention method by the laboratory experiment and analyses and discusses the effect for plant growth from AMD and FA.

ACID MINE DRAINAGE

Mechanics of acid mine drainage generation: AMD is low pH water with high sulfate, high concentrations of iron and other heavy metals occurred by chemical reacting with sulfide mineral containing waste rock, oxygen and water. In case of waste rock, the occurrence of acid water from waste rock is caused by geochemical characteristics associated with the existence of sulfide mineral and physical phenomenon such as weathering process which leads to increase of reactive surface area. The result of oxidation reaction in the generation of acidic drainage can be simply expressed by the following reactions:

$$FeS_2+7/2\ O_2+H_2O \rightarrow Fe^{2+}+2\ SO_4^{2-}+2\ H^+ \quad \text{(a1)}$$

$$Fe^{2+}+1/4\ O_2+H^+ \rightarrow Fe^{3+}+1/2\ H_2O \quad \text{(a2)}$$

$$Fe^{3+}+3\ H_2O+ \rightarrow Fe\ (OH)_{3(s)}+3\ H^+ \quad \text{(a3)}$$

$$FeS_2+14\ Fe^{3+}+8\ H_2O \rightarrow 15\ Fe^{2+}+ 2SO_4^{2-}+16\ H^+ \quad \text{(a4)}$$

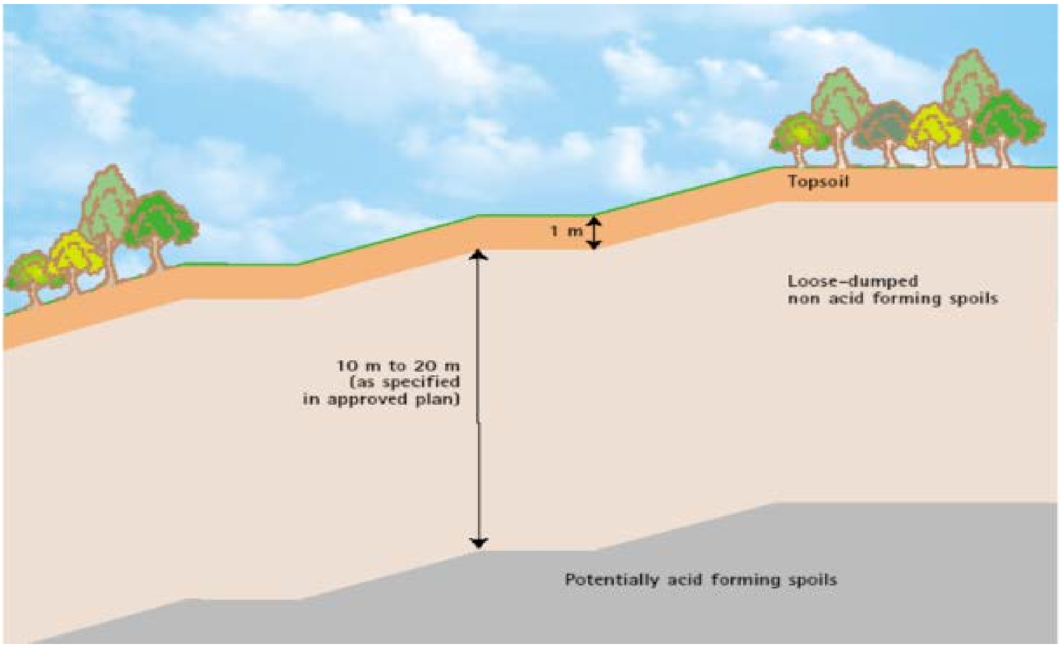
As the first step, $Fe^{2+}$and sulfate ion are generated by oxidized with oxygen and pyrite containing waste rock (a1). If the surrounding environment is sufficiently oxidizing, much of the ferrous iron oxidizes to ferric ion (a2). At pH values above 2.3-3.5, the ferric ion precipitates as Fe $(OH)_3$, leaving little Fe^{3+} in solution whereas lowering the pH at the same time (a3). Any Fe^{3+} that does not precipitate from solution may be used to oxidize additional pyrite (a4) (Nugraha *et al*., 2008).

From these reactions, it can be noted that H^+, sulfate ion, iron ion and water controls the water quality. Furthermore, in terms of physical phenomena, water plays an important role in acid mine drainage (Bussière *et al*., 2004). On non-durable slaking rock material such as coal measure rock, physical weathering that initially induced by wet and dry cycles causes the deterioration of the rock and consequently decreasing the particle size of rock matrix (Birkeland, 1999). This condition increases the reactive surface area and further accelerates the chemical process, both the oxidation rate of sulfide and the neutralizing reaction (Davis and Ritchie, 1987). On the other hand, from physical point of view, a finer-grained material, in spite of resulting low permeability due to small pore size, has also a much higher water retention characteristic than the coarse-grained rock (Devasahayam, 2007). The existence of water in the pore space controls the oxygen diffusion, slaking and the reaction rate, both of sulfide oxidation and alkali dissolution from the minerals contained in rock (Gosselin *et al*., 2007).

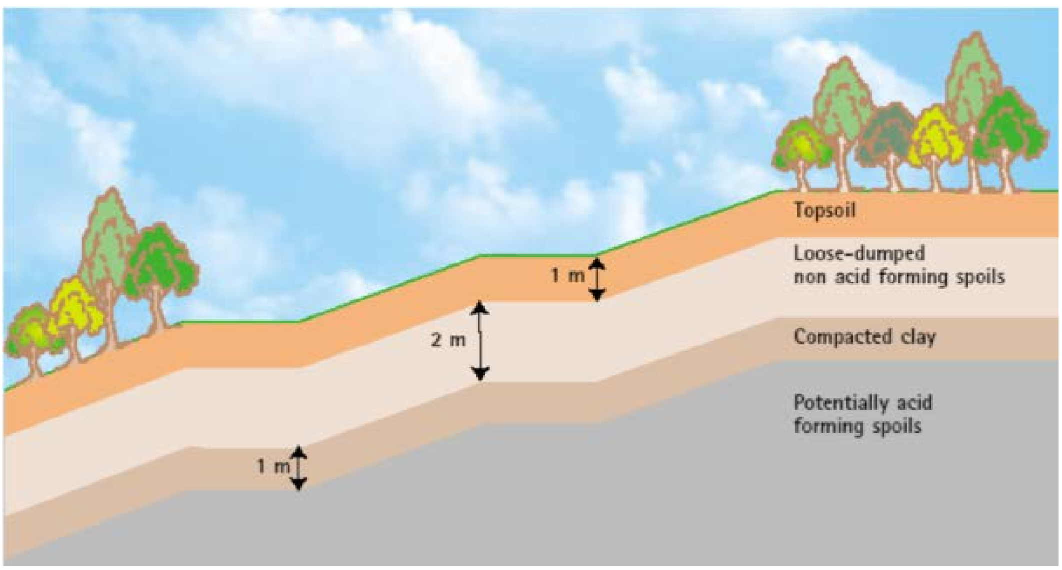
At the surface layer of uncovered waste rock dump, rocks interacts with climate components involving rainfall, solar light, temperature and humidity, which controls the rock weathering and water balance in the surface material. Considering the average temperature and humidity are relatively same *in-situ*, it can be expected that the amount and interval of rainfall is the biggest factor that determines the wetting and drying cycles of surface layer and affects to the weathering rate and water balance of material.

Control of Acid Mine Drainage by the cover layer system: Practices on AMD management on prevention have been implemented on Indonesian coal mine. In the PT Kaltim Prima Coal as one of the biggest coal mine in Indonesia, the most comprehensive effort on the dry cover system is conducted.

AMD prevention by the dry cover system is conducted through the overburden management that achieved by selective dumping to form a cover system. There are some processes to apply this kind of system since exploration stage up to mining operation stage. First, geochemical characterization of waste rock is conducted to divide Non Acid Forming (NAF) and Potentially Acid Forming (PAF) by determining acid producing capacity and acid neutralizing capacity by such kind of ABA and NAG test. Once a rock sample is classified as PAF or NAF based on a certain criteria from geochemical characteristics determined by these tests, a mining company can decide the more detailed plan for the modeling of the mining area. The constructed block model is then used as guidance for a mine planning to develop day to day overburden excavation and dumping strategy to achieve selective dumping that has been planned by encapsulating the PAF with the NAF rock. The encapsulation scenario is depending on the available composition of PAF and NAF material in the mining sequence. When there are enough amounts of the NAF material in the mining



(a): Loose NAF cover



(b): Compacted cover

Fig. 2: Dry cover system implemented at KPC coal mine

area, the Loose NAF cover is preferred in principle due to more cost effective, low erosion risk and more geotechnical stable. In other cases, the construction of the compacted layer of 2 m NAF or 1 m clay to form a barrier to oxygen diffusion is recommended (Fig. 2) (Nugraha *et al*., 2007).

Other method for AMD prevention is discussed the utilization of the FA as the acid neutralizing material. Kusuma *et al*. (2011) suggested the utilization of coal combustion ash as an alternative material of waste rock for minimizing AMD generation in the waste rock dump because it has the high acid neutralizing capacity potential. However, FAmay cause the pollution of leachate and give some impacts for surrounding environment because it usually contains the some kinds of heavy metals that leads to ecological damages. Therefore, an appropriate management and usage of FA have to be considered in order to neutralize the AMD.

Effect for the vegetation: In general, the ground conditions of disturbed land is very severe for

Table 1: Standard in metal ion

Metal ion	Effluent standard in Indonesia (ppm)
Al	-
Cr	5
As	5
Se	1
Cd	1

revegetation/planting. Before starting the revegetation process, the topsoil and subsoil have to be placed above the overburden in order to provide a suitable environment for plant growth. Where the underlying materials have adverse characteristics for root growth, the more depth of topsoil has to be prepared in order to avoid the impact from the adverse material to root growth. However, the enough depth of the topsoil layer cannot prepare in some cases due to the lack of topsoil and subsoil. Therefore, the effect for the vegetation from underlying materials should be considered and evaluated correctly before the revegetation stage because the more cost and labor are needed to repair and maintenance the ground when any problems discover in this stage.

AMD can be a considerable impact for plant growth. Acidity inhibits growth by causing ion toxicity or deficiencies to the plant by releasing or holding certain ions. The productivity of the plants and the number of species are sharply reduced due to highly acidic conditions even though some plants can grow under such conditions. In copper mine in Galicia of Spain, the survival and growth rates of planting show significantly lower under the low pH and high levels of heavy metals and of toxic Al (Álvarez *et al*., 2010; Conesa *et al*., 2006). Therefore, an appropriate evaluation of the characteristics of waste rock has to be needed because the significant impacts on plant growth can be expected by the elution of toxic ions under the acidic condition. Moreover, when the FA is applied to the overburden as the acid neutralizing material, dissolution of heavy metals and trace elements also have to be considered for minimizing the environmental impacts (Table 1).

MATERIAL AND METHODS

Waste rock characterization: Geochemical Static tests were conducted to estimate the potential of AMD generation. The tests were consisted of Net Acid Generation (NAG) test and Acid Base Accounting (ABA) analysis including Acid Neutralizing Capacity (ANC) test and Net Acid Producing Potential (NAPP) calculation. NAG test was conducted by adding 250 mL of 15% hydrogen peroxide (H_2O_2) to 2.5 g of the sample. Hydrogen peroxide was allowed to react with the rock sample overnight. At the following day, the sample was gently heated to accelerate the oxidation of sulfide minerals. Next, the sample and peroxide were boiled for several minutes to evaporate off residual peroxide. After cooling to room temperature, titration by standardized NaOH was carried out to pH = 4.5 and continued to pH = 7. Amount of NaOH titrated that indicates the acidity of solution used to estimate the net amount of acid produced per unit weight of sample expressed in unit of kg H_2SO_4/ton.

ABA analysis defines the amounts and relative balance of potentially acid generating and acid neutralizing minerals in the rock samples by calculating NAPP. While amounts of potentially acid generating is quantified by Acid Potential (AP) calculated from sulfur contents gained from X-ray fluorescence (XRF) analysis, amounts of potentially acid-neutralizing is quantified by ANC calculated from the amount of acid titrated into the reagent reacted with rock sample to neutralize.

ANC is used to quantify the inherent acid buffering capacity of sample regarding to acid neutralizing minerals contained within the sample. This method involves the addition of a known amount of standardized hydrochloric acid to an accurately weighed sample, allowing the sample time to react (with heating) for 1 to 2 h, then back-titrating the mixture with standardized sodium hydroxide to determine the amount of un-reacted hydrochloric acid. The amount of acid consumed by reaction with the sample is then calculated as ANC expressed in unit of kg H_2SO_4/ton. AP is used to quantify the maximum amount of acid potentially generated by presuming sulfur contained in the rock as sulfide minerals to produce AMD. Sulfur contents are obtained from X-ray fluorescence (XRF) analysis by a Rigaku RIX 3100 X-ray fluorescence spectrometer. ANC, AP and NAPP are expressed in units of kgH_2SO_4/ton and calculated as follows and the results of geochemical analysis of sample are shown in Table 2:

$$ANC = [(A\times B)-(C\times D)]/W\times 49 \quad (1)$$

$$AP = 30.6\times S \quad (2)$$

$$NAPP = AP-ANC \quad (3)$$

where,

ANC = Acid Neutralizing Capacity (kg H_2SO_4/ton of material)
A = Concentration of HCl (M)
B = Volume of HCl added (mL)
C = Concentration of NaOH (M)
D = Volume of NaOH used in the titration (mL)
W = The weight of rock sample (g)
AP = Acid Potential Capacity (kg H_2SO_4/ton of material)
S = Sulfur content (%)
NAPP = Net Acid Producing Potential (kg H_2SO_4/ton of material)

Column test: Column with diameter of 50 mm and 150 mm in height were used in the leaching experiment. This column was equipped with a piece of

Table 2: Geochemical characteristics of the samples

Sample	NAF	FA	PAF
NAG pH	3.85	7.57	2.45
AP (kg H_2SO_4/ton)	27.9	0	129.64
ANC (kg H_2SO_4/ton)	41.9	62.475	0
NAPP (kg H_2SO_4/ton)	-14	-62.475	129.64

Table 3: Column filling pattern

	Sample volume rate (%)		
No.	NAF	FA	PAF
1	100	0	0
2	0	100	0
3	0	0	100
4	90	0	10
5	80	0	20
6	70	0	30
7	0	90	10
8	0	80	20
9	0	70	30

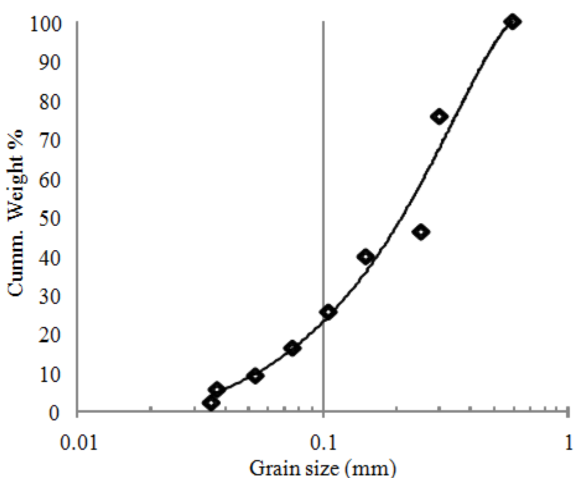
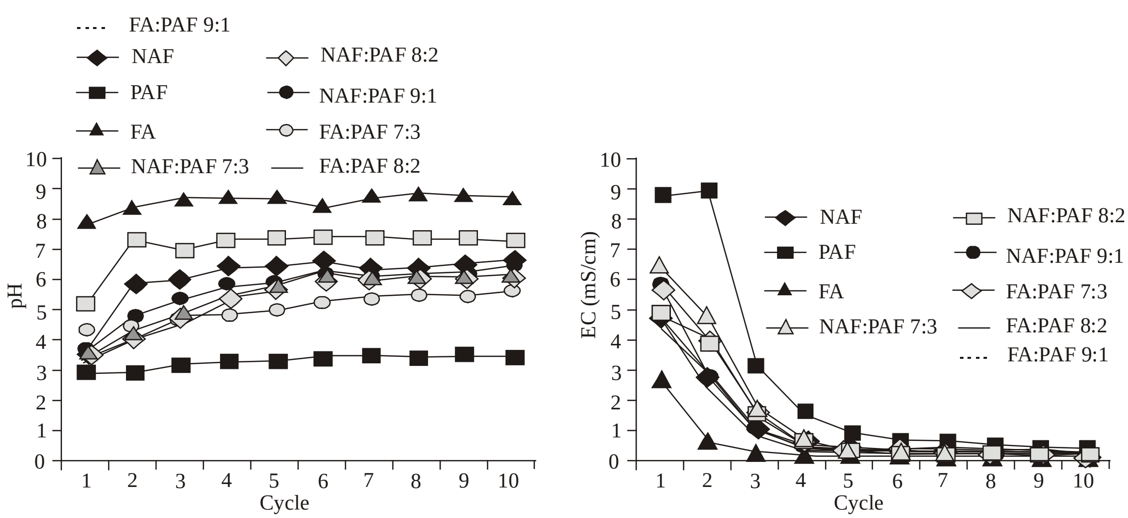


Fig. 3: Particle size distribution of FA

filter paper which installed at the bottom of sample to filter the effluent. Six columns with mixing layer of the materials to simulate the scenario of covering system and other three control's columns were prepared (Table 3). Particle size diameter of the NAF and PAF material used was between 1 to 2 cm and that of FA shows the Fig. 3. Amount of each material needed in the column to perform this test was determined based on volume percentages. Flushing was conducted every day by using de-ionized water of 150 mL and continued 10 cycles. Consideration of the number of 10 cycles test was based on the experience of the column tests conducted in the previous research which the steady state was reached in the seventh cycle (Shimada *et al*., 2012). In addition, this number of cycles was in accordance with the minimum number of test cycles suggested by Sobek *et al*. (1978). Leachate from the columns following daily flushing were collected and sampled which was then measured for pH and Electrical Conductivity (EC). The metal ion contents such as aluminum, chromium, arsenic, selenium and cadmium was analyzed in selected cycle from each column after filtered through 0.45 mm filters. The pH and EC were measured TOA-DK pH meter HM-21P series and HORIBA conductivity meter B-173, respectively. The metal ion contents were measured by using ICP-MS Agilent 7500 Series.

RESULTS AND DISCUSSION

pH and electrical conductivity: The control columns of NAF and FA produce leachate with high pH values, whereas the PAF column has the lowest pH values while the pH value of leachate from the mixing layer columns with the different compositions are between pH values of NAF, FA and PAF (Fig. 4a).



(a): pH

(b): EC

Fig. 4: pH and electrical conductivity of leachate

Table 4: Results of leachate in 1st and 6th cycle

1st cycle

	NAF	FA	PAF	NAF:PAF 9:1	NAF:PAF 8:2	NAF:PAF 7:3	FA:PAF 9:1	FA:PAF 8:2	FA:PAF 8:2
pH	3.7	7.9	2.9	5.2	4.4	4.3	3.7	3.5	3.3
EC (mS/cm)	4.6	2.6	8.7	5.8	4.7	6.3	5.8	4.3	4.5
Fe (ppm)	330.60	9.77	1308.69	408.47	371.88	508.50	11.78	17.84	43.72
Al (ppm)	21.33	5.81	1415.65	112.54	191.47	402.59	19.85	204.72	470.35
Cr (ppm)	0.112	0.041	2.316	0.207	0.352	0.626	0.062	0.178	0.373
As (ppm)	0.073	0.078	0.685	0.013	0.039	0.065	0.017	0.069	0.056
Se (ppm)	0.155	0.117	0.029	0.130	0.054	0.063	0.134	0.138	0.109
Cd (ppm)	0.019	0.002	0.059	0.017	0.013	0.026	0.006	0.012	0.021

6th cycle

	NAF	FA	PAF	NAF:PAF 9:1	NAF:PAF 8:2	NAF:PAF 7:3	FA:PAF 9:1	FA:PAF 8:2	FA:PAF 8:2
pH	6.5	8.4	3.4	6.3	6.1	6.1	7.4	6.4	5.3
EC (mS/cm)	0.300	0.085	0.630	0.270	0.196	0.200	0.115	0.122	0.165
Fe (ppm)	11.47	6.36	21.16	7.610	12.08	16.57	7.530	8.800	6.960
Al (ppm)	74.40	2.00	10.78	3.020	34.25	33.50	2.290	11.00	9.140
Cr (ppm)	0.041	0.054	0.033	0.021	0.050	0.050	0.041	0.058	0.075
As (ppm)	0.090	0.116	0.017	0.013	0.004	0.017	0.065	0.004	0.004
Se (ppm)	0.075	0.109	0.029	0.084	0.033	0.109	0.138	0.096	0.100
Cd (ppm)	0.007	0.003	0.002	0.001	0.002	0.001	0.003	N.D	0.002

The pH of the FA column is the highest (around pH = 8.0) with a downward trend showing the decrease of the dissolution rate as well as the availability of alkaline elements in the column. At the NAF column, after the pH value increased from 3.7 to 6.4 by the fourth cycle, the pH value keeps the stable value in the range of 6.5. Relatively low pH value at the beginning cycle is in accordance with the presence of residual acid and the dissolved acid by weathering products, which cannot be balanced by the dissolution rate of alkaline minerals in the sample to neutralize the acid. The PAF column shows the lowest pH value in any cycle.

The pH of the mixing layer columns improves during the experiment as well as NAF column. In the mixing layer columns, it can be expected that the carbonate minerals or alkali minerals contained in NAF and FA functioned as alkaline donors that consume the acid released by PAF material. The EC of leachate decreases rapidly by the sixth cycle in the range 9 to 1 mS/cm then continue to decrease slightly keeping low values until the end of experiment (Fig. 4b). From these results, it can be said that AMD prevention can be implemented efficiently by mixing PAF with NAF or FA in the appropriate mixing rate.

Metal ion concentration: The understanding of the dissolution of metal ions from waste rock and FA is the considerable topic in order to discuss the effect for plant growing in the revegetation process. The solubilization of aluminum and the excess of the heavy metals cause inhibition of plant growing. The aluminum is accelerated to dissolve as a metal ion under the low pH. FA contains some kinds of the heavy metals though its properties depend on the coal qualities and combustion conditions. The water analysis results in 1st and 6th cycle show the Table 4.

In the PAF column of 1st cycle, the high concentration of Al, Cr and As can be observed. The mixing column with 30% NAF material also shows same tendency with the PAF column. Considering the both columns show pH = 3 approximately, it can be estimated that the excess of metal contents dissolve under the strong acidity condition and the dissolution of metal ions gives the adverse effect for the plant growth. The concentration of heavy metal ions from FA is not so high compared with other columns though that of As and Se shows relatively high in the 6^{th} cycle. Moreover, the concentrations of the metal contents of all columns in 6^{th} cycle are very low compared to that of 1st cycle and the effluent standard in Indonesia (Table 1) since the high pH circumstance reduces the solubility of metals. This finding also means that the impact to plant growth by metal ion contents eluted from the underlying materials can be minimized by effective AMD prevention and preparing an appropriate curing period of the ground until the potential of dissolution of metal ions are decreased. Therefore, the appropriate management and preventative method of AMD are important not only for surrounding environmental protections but also conducting revegetation in the post mined area.

CONCLUSION

The adequate rehabilitation program has to be established in Indonesian coal mining because almost of the coal resources are produced from open-cut mines in Indonesia and the large scale development of its mines causes many kinds of environmental problems. AMD is considered as one of the most significant environmental issues in most coal surface mines caused by the sulphide contents contained in waste rocks and a great deal of rainfall. The appropriate prevention and/or management of the AMD have to be needed for the success of rehabilitation of the post mined land.

In this research, the applicability of dry cover systems including the utilization of FA was discussed for the preventive management of acid mine drainage considering the effect for the vegetation. As a result, it can be understood that to prevent and/or manage the AMD has an important role not only for surrounding environmental protections but also conducting revegetation in the post mined land. However, the more detailed researches have to be implemented for the proper AMD management and the effect for vegetation from cover layer such as time degradation of cover layer, the influence of the different reactive surface area by the different particle size, the properties of FA and the selecting plant species with a high tolerance to excess metal concentrations.

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